

Possible evidence of non-Fermi liquid behavior from quasi-one-dimensional indium nanowires

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We report possible evidence of non-Fermi liquid (NFL) observed at room temperature from the quasi one-dimensional (1D) indium (In) nanowires self-assembled on Si(111)- 7×7 surface. Using high-resolution electron-energy-loss spectroscopy, we have measured energy and width dispersions of a low energy intrasubband plasmon excitation in the In nanowires. We observe the energy-momentum dispersion $\omega(q)$ in the low q limit exactly as predicted by both NFL theory and the random-phase-approximation. The unusual non-analytic width dispersion $\zeta(q) \sim q^\alpha$ measured with an exponent $\alpha=1.40\pm 0.24$, however, is understood only by the NFL theory. Such an abnormal width dispersion of low energy excitations may probe the NFL feature of a non-ideal 1D interacting electron system despite the significantly suppressed spin-charge separation (≤ 40 meV).

I. INTRODUCTION

Since the early prediction of non-Fermi liquid (NFL) behavior for an one-dimensional (1D) interacting electron system,[1] numerous efforts have been made to find evidence of NFL from various forms of 1D conductors including the earlier fractional quantum Hall system,[2] carbon nanotubes,[3] self-assembled nanowires on solid surfaces,[4–6] and anisotropic bulk materials.[7–10] The essential nature of NFL in the 1D electrons systems with enhanced electron correlations is the low energy bosonic collective excitations rather than the single-particle excitations in Fermi liquids.[11] Photoemission spectroscopy (PES) has been used most extensively to probe characteristic features of NFL such as the power law behaviors of spectral function near Fermi level,[3, 10] the spin-charge separation,[4, 7, 8] and the presence of pseudo-gap.[9] Since any deviation from an ideal 1D electrons system such as the presence of impurities, disorder, and thermal fluctuation may sensitively affect the dynamics of these collective excitations,[10–12] continued discussions have been made for some NFL systems claimed earlier.[4, 5] Although one expects infinite life time for such excitations in an ideal NFL, added interactions due to any deviations in real 1D systems may cause the damping of life time τ . Samokhin showed that the damped life time caused by collisions between the excitations due to the nonlinear band curvature even in a clean NFL system

could manifest itself as the non-analytic dispersion $\zeta(q)$ for the width ($\sim \tau^{-1}$) of a spectral peak stemming from the excitations.[11, 12] Such a peculiar spectral behavior may sensitively probe the NFL nature when other spectroscopic evidence is intriguingly suppressed in pragmatic 1D systems.

High energy-resolution (≤ 3 meV) electron-energy-loss spectroscopy (HREELS) has been used to detect such damped collective excitations, plasmons in particular, in the 1D interacting electron systems.[13–15] In the long wavelength limit, intrasubband plasmon shows an almost linear energy-momentum dispersion $\omega(q)$ as observed experimentally[16] and also predicted theoretically both by a NFL theory such as a Luttinger theory and by a typical nearly free-electron gas theory of the random-phase-approximation (RPA).[17] Despite numerous theoretical[11, 12, 17, 18] and experimental studies[13, 16, 19] on plasmon excitations, no unambiguous clue for the NFL behavior due to the damped life time, however, has been reported to our knowledge.

Here we report evidence of the NFL in the quasi 1D indium (In) nanowires self-assembled on the Si(111)- 7×7 surface at room temperature. We find a width dispersion $\zeta(q) \sim q^\alpha$ with an exponent $\alpha=1.40\pm 0.24$ of the low energy intrasubband plasmon excitation. This non-analytic width dispersion is understood only within the framework of NFL theory, never expected from the RPA.[11–13] Unlike the clean NFL, such a peculiar behavior may arise from the non-ideal features of a real 1D sample including the quasi-1D nature allowing extra conduction channels or localization due to impurities and thermal agitation at finite temperature. It has been, indeed, discussed for any signature of the NFL from the 1D conducting In nanowires at room temperature.[6, 20–24]

Although extra interactions due to such deviations may suppress typical NFL features observed

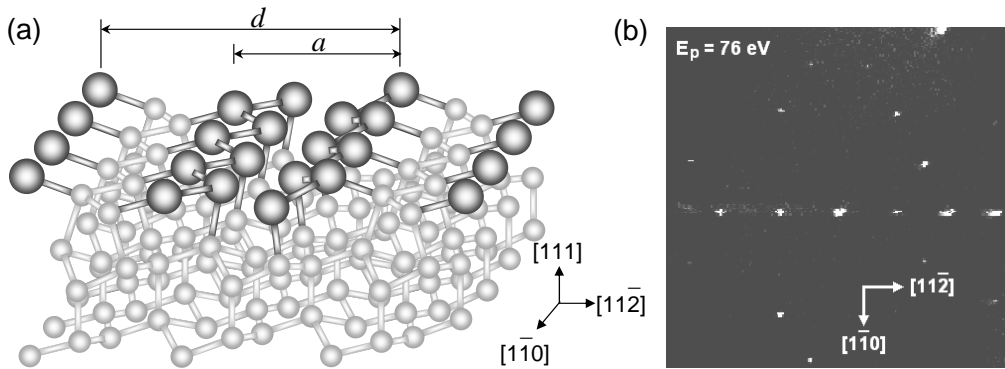


FIG. 1: (a) Atomic arrangement of In nanowires of the In/Si(111)- 4×1 surface. Indium atoms forming quasi 1D nanowires are drawn as dark circles while substrate silicon atoms as grey circles. Four In nanowires form a bundle of width $a=6.9$ Å, and the bundles are separated by $d=13.3$ Å from each other.[20] (b) LEED pattern obtained from the dominant single domain In/Si(111)- 4×1 surface.

in PES studies significantly, the width dispersion, however, appears to survive and exhibits its unique non-analytic dispersion of the NFL even at room temperature.

The atomic arrangement of the In nanowires is depicted in 1(a), where four In nanowires form a metallic bundle along the direction parallel to the nanowires ($[1\bar{1}0]$). [20] Note that the In bundles are arranged periodically along the direction ($[11\bar{2}]$) perpendicular to the length of the bundles. Although the transition from the metallic 4×1 at room temperature to the insulating 8×2 phase below $T_c=120$ K has been understood in terms of Peierls instabilities accompanying the formation of charge-density-wave (CDW) with a doubled periodicity, [6, 24, 25] some observations, for example, the partially suppressed spectral intensity near Fermi level below T_c have not been properly understood. [6, 24] We ascribe such spectral features to the intrinsic nature of the NFL phase.

II. EXPERIMENTS

We have obtained our HREELS data by using a Leybold-Heraeus ELS-22 spectrometer under ultra-high vacuum environment of a base pressure below 1×10^{-10} mbar. The optimum energy resolution and the half-acceptance angle of the detector are 19 meV and 2° , respectively. The dispersion data have been obtained by rotating the sample holder while the monochromator and the analyzer are fixed in position. Indium atoms were deposited onto the Si(111)- 7×7 surface by thermally evaporating the In rod wrapped by a tungsten filament. The single domain 4×1 phase shown in 1(b) has been obtained by controlling the direction of heating current at 400°C . By using a high spatial resolution low energy electron diffraction (SPA-LEED), we estimate the single domains covering the sample surface more than 93%. No vibrational loss peaks associated with contamination have been detected during the measurements.

III. RESULTS AND DISCUSSIONS

We present angle-resolved EEL spectra in 2 with the wave vector (q) parallel (a) and perpendicular (b) to the length of In nanowire. The parallel component of the wave vector q_{\parallel} is determined by

$$q_{\parallel} = \sqrt{\frac{2m}{\hbar^2}} [(E_p - \hbar\omega)^{1/2} \sin \theta_s - E_p^{1/2} \sin \theta_i] \quad (1)$$

where E_p is primary electron energy and $\hbar\omega$ is loss energy. θ_s (θ_i) is scattered (incident) angle of the electron beam. The spectra have been fitted to determine values of energy and width of the

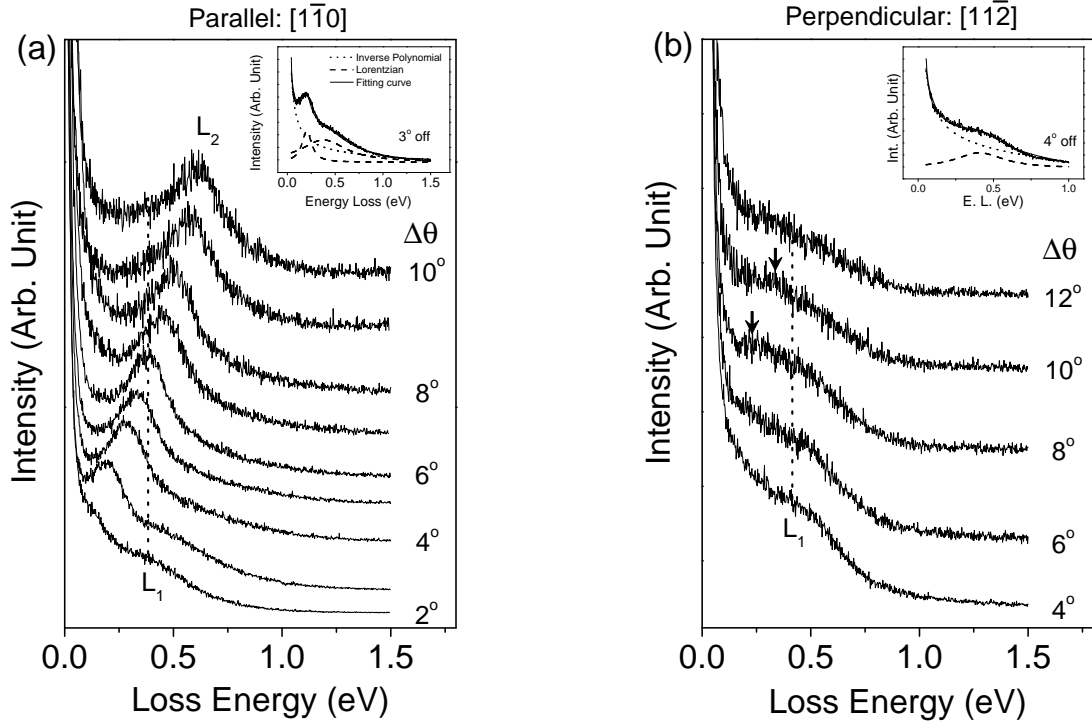


FIG. 2: Angle-resolved EELS spectra obtained from the In/Si(111)- 4×1 surface along the parallel (a) and perpendicular (b) directions to the In nanowires. All spectra are normalized and the primary electron energy is 5.0 eV. $\Delta\theta$ is the angle off from the specular geometry. In (a) two loss peaks of Lorentzian shape, L_1 (dashed vertical line) and L_2 , are found from best fit (see inset) of the spectra while only one non-dispersive L_1 is found in (b). The peaks marked by arrows in (b) are of the same origin with the L_2 from minor domains (see text).

loss peaks as a function of q . Inset in 2 shows the Lorentzian fit functions (dashed curves) and the inverse polynomial background (dotted curves) subtracted from the raw spectra as done earlier.[26] The resulting fit-curves (solid curves) are superimposed on the data revealing excellent fits.

One finds two loss peaks in 2(a), a weak non-dispersive L_1 peak of loss energy centered at 378 ± 11 meV and another quite dispersive L_2 peak along the parallel direction. Note that there exists only L_1 along the direction perpendicular to the In nanowires. The non-dispersive L_1 is ascribed to an interband transition between the three surface bands m_1 , m_2 , and m_3 found in photoemission near the Fermi level, because they are nearly parallel each other with a separation of ~ 350 meV.[6, 22] This interband transition then has to be visible along both directions parallel and perpendicular to the In nanowires. The significant anisotropic dispersions shown in 2 nicely demonstrate the 1D character of the In nanowires. The loss peaks marked by arrows in the spectra

of 8° and 10° in 2(b) are of the L_2 origin stemming from the minor rotational domains due to the 120° rotational symmetry of a Si(111) surface as discussed later. The strongly dispersive peak L_2 is unique only in the metallic 4×1 phase since it is absent both in the clean Si(111)- 7×7 and in the In/Si(111)- $\sqrt{31}\times\sqrt{31}$.

One may think of several possible origins for L_2 ; an interband transition, a local atomic vibration, and a collective excitation such as plasmon or surface phonon. Since the three surface bands near the Fermi level are quite parallel to each other,[6, 22] no dispersive interband transition is allowed for the 4×1 phase. The relatively broad linewidth of L_2 (≥ 137 meV) rules out the local vibrational origin of width typically less than 25 meV. Since the loss energy of L_2 (up to 619 ± 23 meV) far exceeds the highest energy of optical phonon of Si crystal (~ 60 meV) in addition to the absence of multiple phonon peaks, one may safely consider L_2 as a plasmon due to the collective excitation of conduction electrons along the nanowires. Furthermore this peak should not be associated with the well known Peierls instability of a 1D metallic system since the range of momentum measured ($0 \text{ \AA}^{-1} \leq q \leq 0.034 \text{ \AA}^{-1}$) is far from the wave vectors $2k_F=0.82 \text{ \AA}^{-1}$ for the intrinsic Peierls instability[6] or that of the Landau damping where plasmon can decay directly into single particle-hole excitations as will be discussed later.[13, 14]. Therefore the plasmon observed is not affected by such singularities.[17]

We then compare our experimental energy dispersion curves $\omega(q)$ of the plasmon to the one predicted by the NFL theory, which is given below for a 1D interacting electron system near Fermi level.[1]

$$\omega(q) = q[v_F^2 + \frac{2}{\pi\hbar}v_F V(q)]^{1/2}, \quad (2)$$

where

$$V(q) = \frac{2e^2}{4\pi\epsilon} K_0(qa). \quad (3)$$

$v_F=\hbar k_F/m^*$ is the 1D Fermi velocity with effective mass m^* , $V(q)$ is the Coulomb potential, and K_0 is the modified Bessel function of the second kind. Note that the spin excitation alone gives only the first term in 2 so that the spin-charge separation is due essentially to the second term when $V(q)\neq 0$. Incidentally one finds that the RPA for a 1D nearly free-electron gas gives the same q -dependence as in 2 in the small q limit.[17] Therefore the energy dispersion $\omega(q)$ alone would not distinguish the NFL from the RPA.

Using $a=6.9 \text{ \AA}$, we have fitted our energy dispersion data with 2 floating m^* as a fitting parameter. The results depicted in 3(a) exhibits charge (solid) and spin (dashed) dispersions with

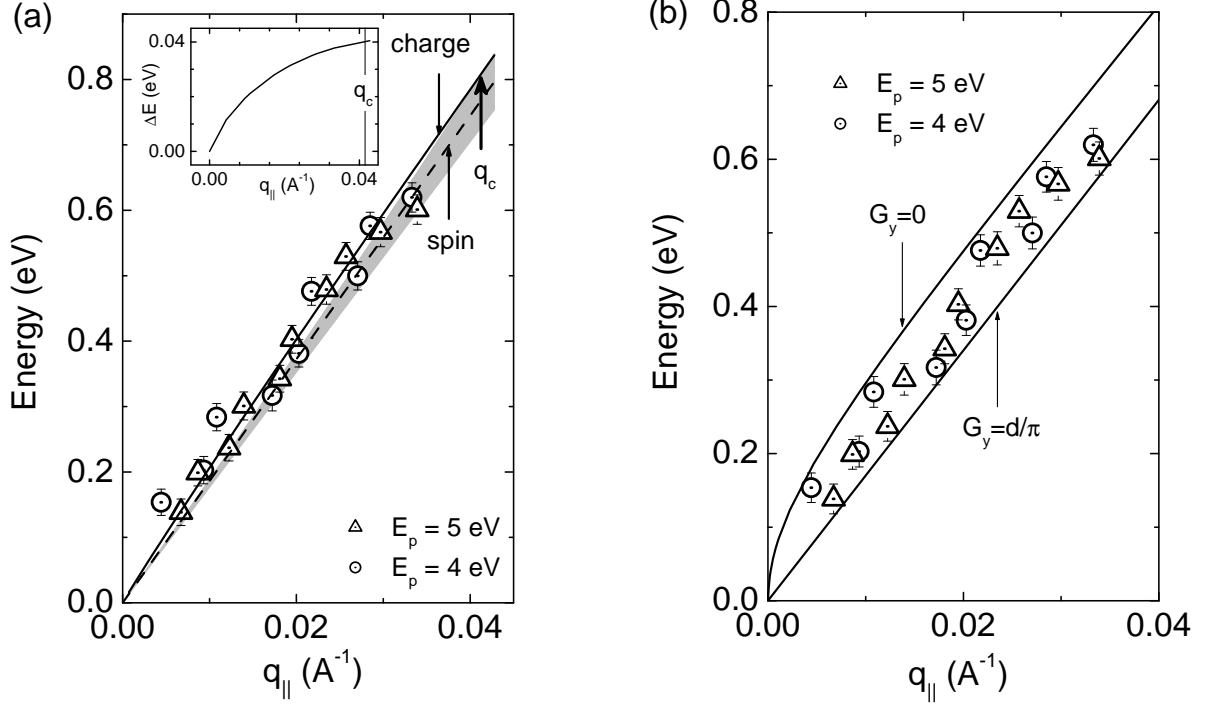


FIG. 3: Fitting the loss energy versus momentum data (empty circles and triangles) with a theoretical formula $\omega(q)$ in 2 without (a) and with (b) inclusion of inter-In bundle interactions. In (a), the theory accounts for our data quite well although the spin (dashed) and charge (solid) separation ΔE is relatively small (≤ 40 meV, see inset). Shaded area corresponds to the particle-hole excitation continuum. In (b), all the data points for $q \leq q_c$ are found within the two limiting dispersions for $G_y=0$ and π/d when inter-bundle interactions are included.

a maximum spin-charge separation of about 40 meV (see inset). One also notes that all our data points locate below the estimated Landau edge $q_c=0.041 \text{ \AA}^{-1}$ determined by the crossing q -value between the charge dispersion and the upper edge of the single particle-hole excitation.[17] Naturally the two dispersions merge to $\omega=0$ as q approaches 0. The spin dispersion found within the single particle-hole excitation and the relatively small spin-charge separation ($\Delta E \leq 40$ meV) may explain why previous photoemission study with energy resolution greater than 100 meV has failed to detect any spin-charge separation at room temperature.

Despite the hidden evidence of NFL in previous photoemission study, [6] some observations still challenge a possibility for the NFL behavior of this 1D metal system. Unlike the bands m_2 and m_3 showing a typical Fermi liquid behavior, the band m_1 , despite its typical Fermi edge at room temperature, becomes severely quenched in spectral intensity at low temperature similar to

the NFL behavior of the carbon nanotubes.[3] Moreover, the asymmetric parameter 0.09 obtained for m_1 in reference [6] remains unaltered despite the phase transition upon cooling, which is not properly understood in terms of either CDW mechanism or metal-insulator transition of Fermi liquids. Previous EELS study challenges no CDW gap or a possibility of partly metallic phase at low temperature.[23] Another theory paper even predicts a metallic surface due to m_1 band for both above and below T_c partially supporting the idea suggested by the EELS study.[24] Such a so-called pseudo-gap feature suggested by the significantly depressed spectral intensity near the Fermi level of an interacting 1D electrons system may be another clue for NFL as for the TTF-TCNQ (tetrathiafulvalene-tetracyanoquinodimethane).[9]

Now the effective masses m^* obtained from the best fits for each of the three parallel surface bands m_1 , m_2 , and m_3 are $m^*/m \sim 0.025$, 0.11, and 0.17, where m is the mass of a free electron. The Fermi wave vectors k_F associated with these are found to be 0.75, 0.54, and 0.41 \AA^{-1} , respectively.[22] The 1D nature of the L_2 is seen also from the two peaks marked by arrows in 2(b) originated from minor domains rotated by $\pm 60^\circ$ from $[1\bar{1}0]$ direction. When the data points for these loss peaks are projected onto the wire direction of the single domain by multiplying $\cos(60^\circ)$ to q , they appear to fall onto the same dispersion curve of the L_2 in 3(a).

We now consider a so-called “band effect” by including the inter-bundle interactions along the $[11\bar{2}]$ direction. With a separation $d=13.3 \text{ \AA}$ between neighboring bundles, we have modified $V(q)$ by adding the second term in 4 in the long wavelength limit.[17]

$$V(q) = \frac{2e^2}{4\pi\epsilon} \left\{ K_0(qa) + 2 \sum_{s=1}^{\infty} K_0(sq d) \cos(G_y s d) \right\} \quad (4)$$

where G_y is the lattice vector perpendicular to the nanowires. As seen in 3(b), we find all the data points for $q \leq q_c$ are contained within the two limiting dispersion curves for two values $G_y=0$ and π/d . We thus conclude that the loss peak L_2 is an intrasubband plasmon excitation with its energy dispersion well described either by the RPA or the NFL theory for $q \leq q_c$.

Although the energy dispersion $\omega(q)$ shows identical behavior for both NFL and nearly free-electron gas in the low q limit, the width dispersion $\zeta(q) \sim q^\alpha$ of the NFL, however, clearly distinguish the NFL nature from nearly free-electron gas when the exponent α appears to be non-analytic.[11, 12] In order to determine this crucial element α , we have fitted two sets of width values of the L_2 with $\zeta(q) \sim q^\alpha$. The best fit (solid curve) gives $\alpha=1.40 \pm 0.24$ as shown in 4(a). As discussed below, this non-analytic width dispersion, in fact, strongly supports the NFL description of the 1D conducting In nanowires at room temperature.

The width of a plasmon excitation may be changed by various possible causes such as disorder, instrumental resolution, and superposition of multiple bands. However, we have excluded these possibilities, since the surface phase remained unchanged maintaining the single domain ($\geq 93\%$) ordered In/Si(111)- 4×1 phase during the entire course of the measurements done with a fixed instrumental resolution ($\Delta q = 0.0038 \pm 0.0002 \text{ \AA}^{-1}$). This was confirmed by observing both no noticeable spectral changes at a fixed momentum and no loss peaks associated with contamination or local defects before and after measurements. The quite symmetric line-shape of the L_2 peak for all q values obtained by subtracting the L_1 as well as the background intensity from the raw spectra excludes also the possibility of broadening due to the superposition of multiple bands.

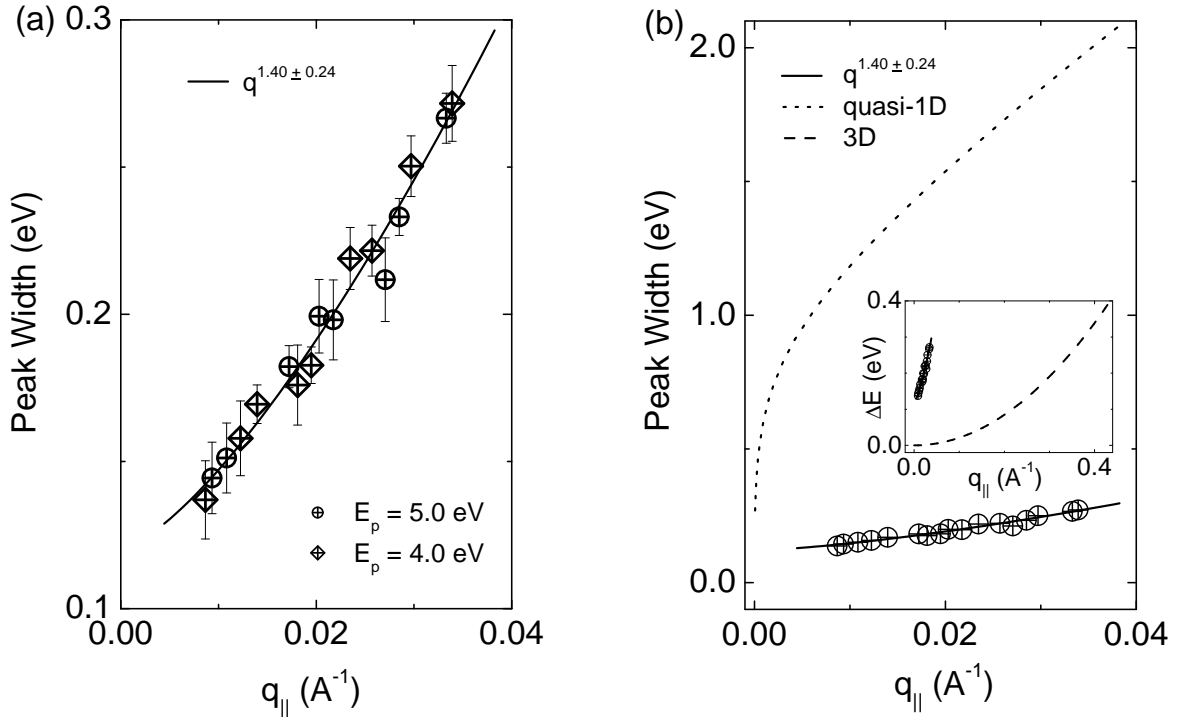


FIG. 4: (a) Results of the fits of our width data (crossed squares and circles) of the intrasubband plasmon with a theoretical fit function $\zeta(q) \sim q^{\alpha}$. The best fit curve (solid curve) gives $\alpha=1.40\pm 0.24$. (b) The estimated dispersions for nearly free-electron gas in quasi-1D (dotted curve) and 3D (dashed curve in inset) electron systems.

We now discuss physical significance of a non-analytic exponent α in real 1D conducting samples. It has been predicted $\alpha=1.5$ for a NFL system with enhanced collisions between bosonic excitations due to non-linear band curvature, which is quite distinct from the clean NFL.[1, 11, 13] This may be quite plausible for the In nanowires since the three surface bands m_1 , m_2 , and m_3 of

the In/Si(111)-4×1 surface have quite non-linear band dispersions. We note also that our width dispersion with $\alpha=1.40\pm0.24$ is clearly distinguished from the non-linear dispersion of quasi-1D nearly free-electron systems where two particle-hole pairs excitations play a vital role.[27] Such a non-linear dispersion is drawn in 4(b) by the dotted curve, which apparently is far from our experimental data. Obviously the dispersion with $\alpha=2$ for volume (3D) plasmons in conventional metals (see inset in 4(b)) may also be safely ruled out since it deviates too far from experimental data.[28] Notice that our width dispersion in 4(a) seems to have a non-zero value as q approaches zero. The finite width at $q=0$ may indicate a significant plasmon life-time decay mainly driven by enhanced Coulomb collision which has never been expected for nearly free-electron gas.[14] The importance of electron correlations for 1D plasmon decay mechanism has recently been observed also for the 1D plasmon from semi-metallic Au chains on the Si(111) surface.[29] However, details of the width dispersion exhibit different behavior mainly due to the lack of the saturation of width beyond $\sim 0.05 \text{ \AA}^{-1}$ as observed from the Au chains,[29] which might originate from relative narrow range $0.004 \text{ \AA}^{-1} \leq q \leq 0.034 \text{ \AA}^{-1}$ of our measurements. Precise measurements at low q range for the Au chains are necessary in order to generalize the intriguing property of elementary excitations in interacting quasi-1D electron systems. The delicate nature of 1D plasmon, however, can appear different when the metallicity of 1D systems are different, i.e., semi-metallic as for the Au chains[30] and metallic as for the In nanowires considered here.

The damping of life time (or equivalently $\zeta^{-1}(q)$) due to thermal fluctuation has also been observed in the single walled carbon nanotubes.[3] Ishii *et al.* reported that single walled carbon nanotubes show the NFL behavior enhanced with decreasing temperature. The finite spectral intensity at Fermi level at room temperature has been interpreted as a result of the NFL features by thermal fluctuation smearing the pseudo gap of the NFL. The reduced spectral intensity near Fermi level observed also for these In nanowires may also be ascribed partially to thermal fluctuation, which may be a signature of the smeared pseudo gap of the NFL phase.[6] Zwick *et al.* provides another example of the NFL phase in the 1D organic conductor TTF-TCNQ where a pseudo gap growing with increasing temperature and the much reduced spectral weight near Fermi level are observed. These are considered as evidence of the NFL phase despite the absence of the spin-charge separation.[9] We thus conclude that the In nanowires, despite their non-ideal quasi 1D nature and finite temperature, reveals the NFL property through the peculiar behavior of the width dispersion of intrasubband plasmon.

IV. CONCLUSION

We have measured the dispersion of an intrasubband plasmon of the quasi-1D In nanowires self-assembled on the Si(111)-7 \times 7 surface at room temperature. We observe quite anisotropic dispersions along the directions parallel and perpendicular to the nanowires demonstrating the 1D nature of the intrasubband plasmon. The non-dispersive EELS peak appearing in both directions is ascribed to an interband transition between three parallel surface bands near Fermi level. The energy dispersion $\omega(q)$ of the unique dispersive peak agrees quite well with predictions by the NFL theory and also by the RPA. The peculiar non-analytic width dispersion of the plasmon, however, as predicted only by the NFL theory strongly supports the NFL nature of the In nanowires even though spin-charge separation is not significant. One may observe similar non-analytic width dispersion of low energy excitations indicating the NFL feature in other real 1D conducting systems.

Acknowledgments

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